

SURFACE ENERGY ARISING FROM THE BEHAVIOR OF LIPID MOLECULES IN THE WATER VIA Γ -CONVERGENCE

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ABSTRACT. We show, in the framework of Γ -convergence, that a surface energy of area type arises from a probabilistic model for lipid molecules in water.

*Dedicated to prof. Giuseppe Grioli,
a constant authoritative presence in Mathematical Physics*

1. Introduction

In a recent paper by PELETIER & RÖGER (see [5]) a simple model for a water-lipid system has been proposed. In such a model a molecule of lipid is represented by a two-bead chain: a head bead, which has an hydrophilic behaviour, and a tail bead, which has an hydrophobic behaviour; heads and tails of the same molecule are connected by a spring. The water molecules are represented by a third type of beads.

The state space at the microscopic level is determined by three positions in \mathbb{R}^3 :

$$X_t^i, \quad X_h^i, \quad X_w^j.$$

X_t^i are the positions of the tails, X_h^i are the positions of the heads and X_w^j are the positions of water; here, $i = 1, \dots, N_\ell$ and $j = 1, \dots, N_w$, being N_ℓ and N_w two fixed integers. Assuming that the beads are confined in a big set $\Omega \subset \mathbb{R}^3$, we can consider the state space for the system as

$$\mathcal{X} := \Omega^{2N_\ell + N_w}.$$

A microstate of the system turns out to be simply a vector of the form

$$X = (X_t^1, \dots, X_t^{N_\ell}, X_h^1, \dots, X_h^{N_\ell}, X_w^1, \dots, X_w^{N_w}).$$

The system can be described in terms of probabilities on \mathcal{X} by means of a density ψ :

$$\psi \in \mathcal{E}, \quad \mathcal{E} := \left\{ \psi: \mathcal{X} \rightarrow [0, +\infty) : \int_{\mathcal{X}} \psi(X) dX = 1 \right\}.$$

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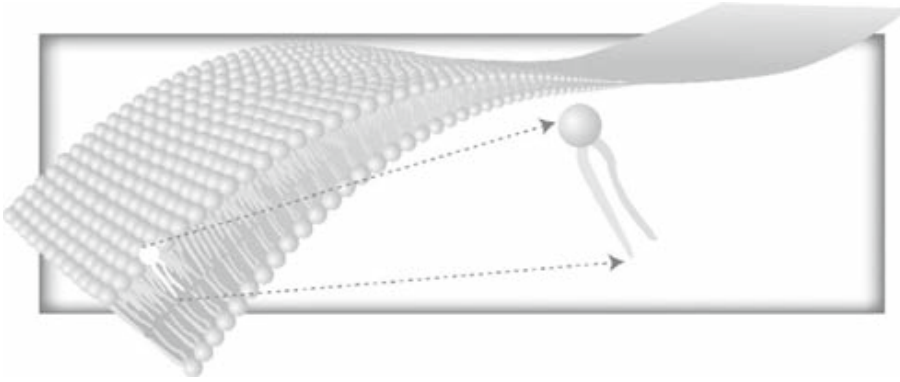


Figure 1. Lipid molecules aggregate into macroscopically surface-like structures

Actually, the observable quantities are three derived quantities, i.e. the volume fractions of tails, heads and water respectively. More precisely, for a given probability density $\psi \in \mathcal{E}$, we denote by $r_h(\psi)(x)$ the probability that in the point $x \in \Omega$ there is an head of a lipid molecule. For tails and water we can define $r_t(\psi)$ and $r_w(\psi)$ in a similar way. The behaviour of the system is governed by two free energies, the sum of an “ideal” free energy and the “non-ideal” one. The ideal free energy wants to represent the interaction between beads of the same molecule, and it penalizes head-tail distance by means of a spring type energy. Regarding this paper, the most important energy we are interested is the non-ideal one, which takes the form

$$\int_{\Omega \times \Omega} (r_w(\psi)(x) + r_h(\psi)(x))r_t(\psi)(y)k(x - y) dx dy$$

being k a convolution kernel: we penalize the proximity of hydrophilic beads (heads and water) and hydrophobic beads (tails). Since it is natural to assume that in any point x of Ω the incompressibility condition

$$r_w(\psi)(x) + r_h(\psi)(x) + r_t(\psi)(x) = 1$$

holds, we immediately get the next expression for the non-ideal energy:

$$\int_{\Omega \times \Omega} (1 - r_t(\psi)(x))r_t(\psi)(y)k(x - y) dx dy.$$

Replacing k by a rescaled version k_δ defined by

$$k_\delta(x) := \delta^{-3}k(x/\delta)$$

in [5] the authors said that one gets the Γ -convergence

$$\frac{2}{\delta} \int_{\Omega \times \Omega} (1 - r_t(\psi)(x))r_t(\psi)(y)k_\delta(x - y) dx dy \xrightarrow{\delta \rightarrow 0} c(k)F^{\text{int}}(r_t(\psi)) \quad (1)$$

where $c(k)$ is a positive constant depending on the function k and

$$F^{\text{int}}(u) := \begin{cases} \mathcal{H}^2(J_u) & u \in BV(\Omega; \{0, 1\}) \\ +\infty & \text{otherwise.} \end{cases}$$

Here, J_u is the discontinuity set of u , so that at the limit we obtain the area of the surface formed by the lipid molecule. Actually, such a convergence result has not been explicitly proved in [5].

The aim of this paper is to investigate convergence (1) proving a rigorous Γ -convergence result; something about the proof can be found in [3].

2. Some preliminary results

In this section we recall some results by ALBERTI & BELLETTINI which are contained in papers [1] and [2].

2.1. An optimal profile problem. In order to state the Γ -convergence result that we will need, we have to investigate first an optimal profile problem.

Let $\rho: \mathbb{R} \rightarrow [0, +\infty)$ be an even function with

$$\int_{-\infty}^{+\infty} \rho(t)(1 + |t|) dt < +\infty$$

and let $W: \mathbb{R} \rightarrow [0, +\infty)$ be a continuous function which vanishes at ± 1 only, and tends to $+\infty$ at infinity. Consider the minimum problem

$$\min_{u \in X} \left\{ \frac{1}{4} \int_{\mathbb{R} \times \mathbb{R}} \rho(t - s)(u(t) - u(s))^2 dt ds + \int_{-\infty}^{+\infty} W(u(t)) dt \right\} \tag{2}$$

where

$$X := \{u: \mathbb{R} \rightarrow [-1, 1] : \lim_{t \rightarrow +\infty} u(t) = +1, \lim_{t \rightarrow -\infty} u(t) = -1\}.$$

It turns out, in [1], that problem (2) has always a solution: we let

$$\sigma(\rho, W) := \min_{u \in X} \left\{ \frac{1}{4} \int_{\mathbb{R} \times \mathbb{R}} \rho(t - s)(u(t) - u(s))^2 dt ds + \int_{-\infty}^{+\infty} W(u(t)) dt \right\}.$$

The next proposition can be found in [1] (see cor. 2.16); in what follows, for any $u \in X$ increasing, the function $u^{-1}: (-1, 1) \rightarrow \mathbb{R}$ is defined by

$$u^{-1}(t) := \inf\{s : t \leq u(s)\}.$$

Proposition 2.1. *Let $f(t) := \max\{-t, 0\}$ and let $g := \rho * f$. For any $u \in X$ increasing and for any $s \in [-1, 1]$ we let*

$$H_u(s) := - \int_{-1}^s \left(\int_s^1 g'(u^{-1}(t')) - u^{-1}(t) \right) dt' dt.$$

Then u solves the problem (2) if and only if $W \geq H_u$ everywhere in $[-1, 1]$ and $W = H_u$ everywhere in the support of the measure $(u^{-1})'$.

By means of proposition 2.1, one is able to solve explicitly the problem (2) for a suitable choice of the potential W , as the next corollary shows.

Corollary 2.2. *Assume that $W(t) \geq c_\rho(1 - t^2)$ everywhere, where*

$$c_\rho := \int_0^{+\infty} \rho(t) dt.$$

Then the function $u(t) := \operatorname{sgn} t$ solves the problem (2). In particular, we have

$$\sigma(\rho, W) = \int_{-\infty}^{+\infty} |t| \rho(t) dt \quad (3)$$

for any admissible ρ .

Proof. By definition, we immediately get $u^{-1}(t) = 0$ everywhere in $(-1, 1)$. Then, for any $s \in [-1, 1]$ we obtain

$$H_u(s) = -g'(0)(1 - s^2).$$

Now,

$$g(t) = \int_{-\infty}^{+\infty} \rho(\tau) f(t - \tau) d\tau$$

hence

$$\begin{aligned} g'(t) &= \int_{-\infty}^{+\infty} \rho(\tau) f'(t - \tau) d\tau = \int_{-\infty}^{+\infty} \rho(t - \tau) f'(\tau) d\tau \\ &= - \int_{-\infty}^0 \rho(t - \tau) d\tau = - \int_t^{+\infty} \rho(r) dr \end{aligned}$$

and then $g'(0) = -c_\rho$, from which we get

$$H_u(s) = c_\rho(1 - s^2) \leq W(s).$$

Moreover, we notice that the support of the measure $(u^{-1})'$ is empty. Applying proposition 2.1 we deduce that u solves problem (2). Finally, (3) is a straightforward computation. \square

2.2. A Γ -convergence result. Let n be a positive integer and let $J: \mathbb{R}^n \rightarrow [0, +\infty)$ be such that $J(x) = J(-x)$ for any $x \in \mathbb{R}^n$ and

$$\int J(x)(1 + |x|) dx < +\infty.$$

For any $\eta > 0$ let $J_\eta: \mathbb{R}^n \rightarrow [0, +\infty)$ be given by

$$J_\eta(x) := \frac{1}{\eta^n} J\left(\frac{x}{\eta}\right).$$

Moreover, let $W: \mathbb{R} \rightarrow [0, +\infty)$ as before, i.e. a continuous function which vanishes at ± 1 only, and tends to $+\infty$ at infinity. Let $\Omega \subset \mathbb{R}^n$ be an open and bounded set with Lipschitz boundary. For any $\varepsilon > 0$ we define $AB_\varepsilon(\cdot, J, W): L^1(\Omega) \rightarrow [0, +\infty]$ as

$$AB_\varepsilon(u, J, W) := \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} J_\varepsilon(x - y)(u(x) - u(y))^2 dx dy + \frac{1}{\varepsilon} \int_{\Omega} W(u(x)) dx.$$

By means of the optimal profile problem investigated in paragraph 2.1 we are able to state the Γ -convergence result for the family $\{AB_\varepsilon(\cdot, J, W)\}_{\varepsilon>0}$ which is contained in [2]. Fix a unit vector $e \in \mathbb{R}^n$. For any $s \in \mathbb{R}$ let

$$J^e(s) := \int_{\{y \in \mathbb{R}^n : \langle y, e \rangle = 0\}} J(y + se) d\mathcal{H}^{n-1}(y).$$

The following Γ -convergence-compactness result holds (see thm 1.4 in [2]).

Theorem 2.3. *Let $AB(\cdot, J, W) : L^1(\Omega) \rightarrow [0, +\infty]$ be given by*

$$AB(u, J, W) := \begin{cases} \int_{J_u} \sigma(J^{\nu_u}, W) d\mathcal{H}^{n-1} & \text{if } u \in BV(\Omega; \{-1, 1\}) \\ +\infty & \text{otherwise in } L^1(\Omega). \end{cases}$$

Then the following conditions hold:

$$\liminf_{j \rightarrow +\infty} AB_{\varepsilon_j}(u_j, J, W) \geq AB(u, J, W), \quad \text{for any } u \in L^1(\Omega), \text{ for any } \varepsilon_j \searrow 0 \tag{4}$$

and for any $u_j \xrightarrow{L^1(\Omega)} u$;

for any $u \in BV(\Omega; \{-1, 1\})$ there exist $\varepsilon_j \searrow 0$ and $u_j \xrightarrow{L^1(\Omega)} u$
with $-1 \leq u_j \leq 1$ a.e. in Ω and with $\limsup_{j \rightarrow +\infty} AB_{\varepsilon_j}(u_j, J, W) \leq AB(u, J, W)$. (5)

In particular, it holds $AB_\varepsilon(\cdot, J, W) \xrightarrow{\Gamma} AB(\cdot, J, W)$ as $\varepsilon \rightarrow 0^+$ with respect to the L^1 -strong topology. Moreover, if (v_j) is a sequence in $L^1(\Omega)$ with $AB_{\varepsilon_j}(v_j) \leq C$ for some positive constant C and some sequence $\varepsilon_j \searrow 0$ then up to subsequence (not relabelled) v_j converges to some $v \in BV(\Omega; \{-1, 1\})$ strongly in $L^1(\Omega)$.

3. Main result

In this section first of all we state a problem which turns out to be the generalization of the problem (1).

Let $k : \mathbb{R} \rightarrow [0, +\infty)$ be a continuous function with compact support. Let n be a positive integer. We let $K : \mathbb{R}^n \rightarrow [0, +\infty)$ as $K(x) := k(|x|)$. For any $\eta > 0$ let $k_\eta : \mathbb{R}^n \rightarrow [0, +\infty)$ be given by

$$k_\eta(x) := \frac{1}{\eta^n} K\left(\frac{x}{\eta}\right).$$

Let $\Omega \subset \mathbb{R}^n$ be an open and bounded set with Lipschitz boundary. For any $\varepsilon > 0$ we define $F_\varepsilon : L^1(\Omega) \rightarrow [0, +\infty]$ as

$$F_\varepsilon(u) := \begin{cases} \frac{2}{\varepsilon} \int_{\Omega \times \Omega} (1 - u(x))u(y)k_\varepsilon(x - y) dx dy & \text{if } 0 \leq u \leq 1 \text{ a.e. in } \Omega \\ +\infty & \text{otherwise in } L^1(\Omega). \end{cases}$$

In what follows we use the notation

$$\omega_n := \mathcal{H}^{n-1}(S^{n-1}), \quad K_{1,n} := \frac{1}{\omega_n} \int_{S^{n-1}} |\langle w, e \rangle| d\mathcal{H}^{n-1}(w), \quad k_n := \int_{\mathbb{R}^n} |x| K(x) dx$$

being e any unit vector in \mathbb{R}^n and $S^{n-1} := \{x \in \mathbb{R}^n : |x| = 1\}$. Moreover, we let

$$\alpha_n := K_{1,n}k_n.$$

The main result of the paper is given by the following theorem.

Theorem 3.1. *It holds $F_\varepsilon \xrightarrow{\Gamma} F$ as $\varepsilon \rightarrow 0^+$ with respect to the L^1 -strong topology, where*

$$F(u) := \begin{cases} \alpha_n \mathcal{H}^{n-1}(J_u) & \text{if } u \in BV(\Omega; \{0, 1\}) \\ +\infty & \text{otherwise in } L^1(\Omega). \end{cases}$$

Moreover, if (u_j) is a sequence in $L^1(\Omega)$ with $F_{\varepsilon_j}(u_j) \leq C$ for some positive constant C and some sequence $\varepsilon_j \searrow 0$ then up to subsequence (not relabelled) u_j converges to some $u \in BV(\Omega; \{0, 1\})$ strongly in $L^1(\Omega)$.

Proof. We divide the proof in several steps.

Step 1. First of all we need to relate F_ε with functional of type AB_ε . We claim that for any $u \in L^1(\Omega)$ with $0 \leq u \leq 1$ a.e. in Ω and for ε sufficiently small we have

$$F_\varepsilon(u) = AB_\varepsilon(2u - 1, K, W_0) \tag{6}$$

where

$$W_0(t) := \frac{\tau_n}{2}|1 - t^2|, \quad \tau_n := \int_{\mathbb{R}^n} K(x) dx.$$

Indeed, let $v := 2u - 1$. Then, $v \in L^1(\Omega)$ and, by direct computation, using the symmetry of K , we have

$$\begin{aligned} F_\varepsilon(u) &= \frac{2}{\varepsilon} \int_{\Omega \times \Omega} (1 - u(x))u(y)k_\varepsilon(x - y) dx dy \\ &= \frac{2}{\varepsilon} \int_{\Omega \times \Omega} \left(1 - \frac{v(x) + 1}{2}\right) \frac{v(y) + 1}{2} k_\varepsilon(x - y) dx dy \\ &= \frac{1}{2\varepsilon} \int_{\Omega \times \Omega} (1 - v(x))(1 + v(y))k_\varepsilon(x - y) dx dy \\ &= \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} (1 - v(x))(1 + v(y))k_\varepsilon(x - y) dx dy \\ &\quad + \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} (1 - v(y))(1 + v(x))k_\varepsilon(x - y) dx dy \\ &= \frac{1}{2\varepsilon} \int_{\Omega \times \Omega} (1 - v(x)v(y))k_\varepsilon(x - y) dx dy \\ &= \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} (2 - 2v(x)v(y) + v^2(x) + v^2(y) - v^2(x) - v^2(y))k_\varepsilon(x - y) dx dy \\ &= \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} k_\varepsilon(x - y)(v(x) - v(y))^2 dx dy + \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} (1 - v^2(x))k_\varepsilon(x - y) dx dy \\ &\quad + \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} (1 - v^2(y))k_\varepsilon(x - y) dx dy. \end{aligned}$$

Now, for any $x \in \Omega$, using the very definition of k_ε , we obtain

$$\int_{\Omega} k_\varepsilon(x - y) dy = \int_{\frac{1}{\varepsilon}(\Omega-x)} K(z) dz.$$

Since k has compact support in \mathbb{R} and Ω is bounded, for ε sufficiently small we get

$$\int_{\frac{1}{\varepsilon}(\Omega-x)} K(z) dz = \int_{\mathbb{R}^n} K(z) dz = \tau_n$$

from which

$$\int_{\Omega \times \Omega} (1 - v^2(x))k_\varepsilon(x - y) dx dy = \tau_n \int_{\Omega} (1 - v^2(x)) dx.$$

In the same way we get

$$\int_{\Omega \times \Omega} (1 - v^2(y))k_\varepsilon(x - y) dx dy = \tau_n \int_{\Omega} (1 - v^2(y)) dy.$$

Therefore we deduce that

$$\begin{aligned} F_\varepsilon(u) &= \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} k_\varepsilon(x - y)(v(x) - v(y))^2 dx dy + \frac{\tau_n}{2\varepsilon} \int_{\Omega} (1 - v^2(x)) dx \\ &= \frac{1}{4\varepsilon} \int_{\Omega \times \Omega} k_\varepsilon(x - y)(v(x) - v(y))^2 dx dy + \frac{1}{\varepsilon} \int_{\Omega} W_0(v(x)) dx \\ &= AB_\varepsilon(v, K, W_0) \\ &= AB_\varepsilon(2u - 1, K, W_0) \end{aligned}$$

and this gives (6).

Step 2. We are ready to prove the compactness of equibounded (in energy) sequences. Let (u_j) be a sequence in $L^1(\Omega)$ with $F_{\varepsilon_j}(u_j) \leq C$ for some positive constant C and some sequence $\varepsilon_j \searrow 0$. Of course we can assume $0 \leq u_j \leq 1$ a.e. in Ω for any $j \in \mathbb{N}$. Let $v_j := 2u_j - 1$. Then $|v_j| \leq 1$ a.e. in Ω and thus, by (6) we get

$$F_{\varepsilon_j}(u_j) = AB_{\varepsilon_j}(v_j, K, W_0).$$

Therefore $AB_{\varepsilon_j}(v_j, K, W_0) \leq C$ and then, by theorem 2.3 we deduce that, up to subsequence not relabelled, v_j converges to some $v \in BV(\Omega; \{-1, 1\})$ strongly in $L^1(\Omega)$. It is sufficient to let $u := \frac{1}{2}(v + 1)$.

Step 3. Next, formula (6) suggests that we have to compute $AB(v, K, W_0)$: we claim that for any $v \in BV(\Omega; \{-1, 1\})$ it holds

$$AB(v, K, W_0) = \alpha_n. \tag{7}$$

It is sufficient to prove that for any $e \in S^{n-1}$ one has $\sigma(K^e, W_0) = \alpha_n$. First of all, we notice that, since K is radially symmetric,

$$\begin{aligned} \int_0^{+\infty} K^e(s) ds &= \int_0^{+\infty} \int_{\{y \in \mathbb{R}^n : \langle y, e \rangle = 0\}} K(y + se) d\mathcal{H}^{n-1}(y) ds \\ &= \int_{\{z \in \mathbb{R}^n : \langle z, e \rangle \geq 0\}} K(z) d\mathcal{H}^{n-1}(z) = \frac{\tau_n}{2}. \end{aligned}$$

Thus, we can apply corollary 2.2 and then we get

$$\sigma(K^e, W_0) = \int_{-\infty}^{+\infty} |t| K^e(t) dt.$$

Now, in order to compute the right hand-side we use the coarea formula and we obtain

$$\begin{aligned} \int_{-\infty}^{+\infty} |t| K^e(t) dt &= \int_{-\infty}^{+\infty} |t| \int_{\{y \in \mathbb{R}^n : \langle y, e \rangle = 0\}} K(y + te) d\mathcal{H}^{n-1}(y) dt \\ &= \int_{-\infty}^{+\infty} |t| \int_{\{z \in \mathbb{R}^n : \langle z, e \rangle = t\}} K(z) d\mathcal{H}^{n-1}(z) dt \\ &= \int_{-\infty}^{+\infty} \int_{\{z \in \mathbb{R}^n : \langle z, e \rangle = t\}} |\langle z, e \rangle| K(z) d\mathcal{H}^{n-1}(z) dt \\ &= \int |\langle x, e \rangle| K(x) dx \\ &= \int_0^{+\infty} \int_{\{y \in \mathbb{R}^n : |y|=r\}} |\langle y, e \rangle| K(y) d\mathcal{H}^{n-1}(y) dr \\ &= \int_0^{+\infty} k(r) \int_{\{y \in \mathbb{R}^n : |y|=r\}} |\langle y, e \rangle| d\mathcal{H}^{n-1}(y) dr. \end{aligned}$$

It is easy to see, for instance using spherical coordinates in \mathbb{R}^n , that

$$\int_{\{y \in \mathbb{R}^n : |y|=r\}} |\langle y, e \rangle| d\mathcal{H}^{n-1}(y) = r^n \int_{S^{n-1}} |\langle w, e \rangle| d\mathcal{H}^{n-1}(w)$$

from which we get, using again the coarea formula,

$$\begin{aligned} \int_0^{+\infty} k(r) \int_{\{y \in \mathbb{R}^n : |y|=r\}} |\langle y, e \rangle| d\mathcal{H}^{n-1}(y) dr &= K_{1,n} \int_0^{+\infty} \omega_n r^n k(r) dr \\ &= K_{1,n} \int_0^{+\infty} \mathcal{H}^{n-1}(S^{n-1}) r^{n-1} r k(r) dr \\ &= K_{1,n} \int_0^{+\infty} \int_{\{x \in \mathbb{R}^n : |x|=r\}} r k(r) d\mathcal{H}^{n-1}(x) dr \\ &= \alpha_n \end{aligned}$$

and this ends the proof of (7).

Step 4. We are ready to prove the Γ -liminf inequality. Let (ε_j) be a positive and infinitesimal sequence and let (u_j) be a sequence in $L^1(\Omega)$ with $u_j \xrightarrow{L^1(\Omega)} u$ for some $u \in L^1(\Omega)$; we have to prove that

$$\liminf_{j \rightarrow +\infty} F_{\varepsilon_j}(u_j) \geq F(u). \quad (8)$$

Of course, we can assume that $0 \leq u_j \leq 1$ a.e. in Ω , otherwise (8) is trivial. Combining (6) with (4) and (7) we obtain, since $2u_j - 1 \xrightarrow{L^1(\Omega)} 2u - 1$,

$$\begin{aligned} \liminf_{j \rightarrow +\infty} F_{\varepsilon_j}(u_j) &= \liminf_{j \rightarrow +\infty} AB_{\varepsilon_j}(2u_j - 1, K, W_0) \geq AB(2u - 1, K, W_0) \\ &= \alpha_n \int_{J_{2u-1}} d\mathcal{H}^{n-1} \\ &= \alpha_n \mathcal{H}^{n-1}(J_u) = F(u) \end{aligned}$$

which is (8).

Step 5. We conclude the proof proving the Γ -limsup inequality. Let $u \in BV(\Omega, \{0, 1\})$ we have to prove that there exist a positive and infinitesimal sequence ε_j and a sequence (u_j) in $L^1(\Omega)$ with $u_j \xrightarrow{L^1(\Omega)} u$ such that

$$\limsup_{j \rightarrow +\infty} F_{\varepsilon_j}(u_j) \leq F(u). \tag{9}$$

Let (ε_j) be a positive and infinitesimal sequence and (v_j) in $L^1(\Omega)$ be such that $v_j \xrightarrow{L^1(\Omega)} 2u - 1$ and such that, conformally to (5),

$$\limsup_{j \rightarrow +\infty} AB_{\varepsilon_j}(v_j, K, W_0) \leq AB(2u - 1, K, W_0) = F(u).$$

Since $|v_j| \leq 1$ we can use again (6) and conclude that, letting $u_j := \frac{1}{2}(v_j + 1)$,

$$\limsup_{j \rightarrow +\infty} F_{\varepsilon_j}(u_j) = \limsup_{j \rightarrow +\infty} AB_{\varepsilon_j}(v_j, K, W_0) \leq F(u)$$

and thus (9) holds. □

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